(12) # EUROPEAN PATENT APPLICATION

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(54) **Packaging bags.**

(57) A packaging bag has an outer layer and an inner layer. The outer layer is a low density ethylene-based polymer or a mixture thereof with an ethylene-vinyl acetate copolymer. The inner layer is a mixture of a high density polyethylene and an ethylene-α-olefin copolymer. This packaging bag has excellent physical properties, e.g. tensile strength and impact resistance, and is suitable for use in the packaging of heavy materials, particularly by an automatic packaging technique.

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TITLE:

Packaging Bags

DESCRIPTION:

The invention relates to packaging bags, and more

5   particularly, to packaging bags which have high stiffness

and blocking resistance and are therefore suitable for

automatic packaging.

Heretofore, heavy-duty bags for packaging granular

materials such as rice and wheat, powdery materials such

10   as fertilizer and feed, and angular solid materials such

as fowl have been made of low density polyethylene.

These heavy-duty bags, however, are limited in their

usefulness because of the low mechanical strength of low

density polyethylene.   In order to compensate for the

15   poor mechanical strength, therefore, it is necessary to

increase the film thickness.   Particularly, in the case

of heavy-duty bags, it is required for the film thick-

ness to be markedly increased because of their seriously

low tensile strength and stiffness.   Furthermore, low

density polyethylene is not suitable for use in the
production of bags for automatic packaging because its
stiffness is poor.

In recent years, therefore, bags made of a laminated

5   film comprising a high density polyethylene layer and a
low density polyethylene layer have been proposed to
overcome the above-described problems of conventional
bags (see Japanese Patent Application Laid-Open Nos
92023/81, 29426/82, and 30994/80).

10   A bag prepared by using a film as disclosed in Japanese
Patent Application Laid-Open Nos. 92023/81 and 29426/82
is very preferred for practical use because the seal
strength and the film impact strength are high, the tear
strength is well balanced, and furthermore, the outer

15   layer is not slippery. However, when the bag is used
to accommodate and convey angular heavy materials such
as fowl, its film puncture strength, tear strength,
impact resistance at low temperatures, and so forth
become problems. Also, a bag made of a film as disclosed

20   in Japanese Patent Application Laid-Open No. 30994/80
suffers from disadvantages in that since the inner layer
of the bag is made of low density polyethylene, blocking
readily occurs and difficulties are encountered in opening
the bag and, furthermore, it takes much labour and time

to place materials in the bag because the inner surface
is slippery.

The invention provides a packaging bag comprising an
outer layer and an inner layer, characterised in that
5   the outer layer is a low density ethylene-based polymer
or a mixture thereof with an ethylene-vinyl acetate
copolymer, and the inner layer is a mixture of a high
density polyethylene and an ethylene-α-olefin copolymer.

The outer layer of the bag of the invention is made of
10   a low density ethylene-based polymer (hereinafter
referred to as "LDPE") or a mixture of LDPE and an
ethylene-vinyl acetate copolymer (hereinafter referred
to as "EVA"). Various types of LDPE can be used in the
invention. Usually they have a density of from 0.91 to
15   0.94 gram per cubic centimeter (g/cm^3), preferably from
0.915 to 0.935 g/cm^3. Their melt indexes (MI) are
within the range of from 0.3 to 10 grams per 10 minutes
(g/10 min), preferably from 0.3 to 4 g/10 min. When MI
is more than 10 g /10 min, the impact resistance is
20   lowered, unsuitably for practical use. On the other
hand, when MI is less than 0.3 g/10 min, processability
becomes low. The LDPE used includes that produced
under high pressure and that produced under low pressure.
Examples of such LDPE produced under low pressure

include copolymers of ethylene and α-olefins containing

from 3 to 12 carbon atoms (e.g. propylene, butene-1,

pentene-1, hexene-1, 4-methylpentene-1 and nonene-1).

The α-olefin unit content of the copolymers is preferably

5   from 1 to 10% by mole.

When the outer layer of the present bag is made of a

mixture of LDPE and EVA, the mixing ratio of LDPE to EVA

is not critical. Usually, however, the ratio of LDPE/EVA

is from 10/90 to 95/5 (by weight), with the range of

10   from 60/40 to 90/10 (by weight) being preferred. If the

proportion of EVA is excessively increased, various

problems arise; for example, a bag produced by using

the resulting resin mixture has acetic acid odour and,

furthermore, the surface of the bag excessively loses its

15   smoothness. Such poor surface smoothness causes

blocking between bags and makes bags stick to conveying

equipments, e.g. a roller and the belt of a belt

conveyor, as used in automatic packaging, resulting in a

serious reduction in workability.

20   The inner layer of the bag of the invention is made of a

mixture of a high density polyethylene (hereinafter

referred to as "HDPE") and an ethylene-α-olefin copolymer.

Various types of HDPE can be used in the invention.

usually they have a density ranging between 0.94 and 0.97

g/cm^3, with the range of from 0.945 to 0.965 g/cm^3 being

preferred. Their melt indexes are within the range of

from 0.01 to 1 g/10 min, preferably from 0.02 to 0.2

g/10 min. When MI is more than 1 g/10 min, the

mechanical strength, e.g. tensile strength, of the

resulting film is undesirably lowered. Various

ethylene-α-olefin copolymers can be used in admixture

with HDPE. The ethylene-α-olefin copolymer suitably has

a density of from 0.910 to 0.940 g/cm^3, preferably from

0.915 to 0.935 g/cm^3, and a MI of from 0.3 to 10 g/10 min,

preferably from 0.3 to 4 g/10 min. Preferred examples

of α-olefins which can be used in the preparation of

the copolymers are those monomers containing from 3

to 12 carbon atoms, such as propylene, butene-1,

pentene-1, hexene-1, 4-methylpentene-1, octene-1 and

nonene-1. The α-olefin unit content of the copolymer

is suitably within the range of from 1 to 10% by mole.

Of these ethylene-α-olefin copolymers, ethylene-

propylene-based copolymer elastomers (hereinafter

referred to as "EP") are most suitable. Various types

of EP can be used in the invention. Those elastomers

having a Mooney viscosity of from 40 to 150 are preferred.

When the Mooney viscosity is less than 40, the mechanical

strength of the resulting film is undesirably lowered.

Preferred examples of EP are ethylene-propylene rubber

and ethylene-propylene terpolymers containing, as a

third component, dienes such as divinylbenzene, 5-

-ethylidene-2-norbornene, 1, 4,-hexadiene,

dicyclopentadiene and cyclooctadiene.

The ratio of HDPE to the ethylene-α-olefin copolymer is
not critical and can be determined appropriately

5   depending on the purpose for which the ultimate bag is
used. Usually, however, the ratio of HDPE to the
ethylene-α-olefin copolymer is from 95/5 to 50/50 (by
weight), with the range of from 90/10 to 60/40 (by weight)
being preferred. When the proportion of the ethylene-α-

10   olefin copolymer is less than 5% by weight, the puncture
strength and seal strength of the resulting film are
reduced, and when a bag prepared by using the film is
dropped, it is easily torn. On the other hand, when it
is more than 50% by weight, processability becomes low,

15   and a bag prepared by using the resulting film has poor
stiffness and is not suitable for practical use.

The bag of the invention comprises the above-described
outer and inner layers which are laminated on each
other. Although the ratio of the thickness of the

20   outer layer to the thickness of the inner layer can be
determined depending on the purpose for which the bag is
used, it is usually determined within the range of 1/99

to 60/40. When the thickness of the outer layer is more
than 60% of the total thickness of the outer and inner
layers, the tensile strength and stiffness of the bag
are undesirably lowered. On the other hand, when it is
5   less than 1%, the processability thereof becomes
difficult.

The bag of the invention is usually of the two layer
laminated structure consisting of an outer layer and an
inner layer. If desired, a three layer laminated
10   structure, including an intermediate layer between the
outer and inner layers or having an innermost layer on
the inner layer, can be used. The type of the
intermediate layer being interposed between the outer
and inner layers is not critical and can be determined
15   appropriately depending on the purpose for which the bag
is used. Although the type of the innermost layer is
not critical as in the case with the intermediate layer,
it is preferably made of an ethylene-α-olefin copolymer.
Although the properties of the ethylene-α-olefin
20   copolymer are not critical, the density is usually from
0.91 to 0.94 g/cm^3 and preferably from 0.915 to 0.935
g/cm^3, and MI is from 0.1 to 20 g/10 min and preferably
from 0.2 to 10 g/10 min. The α-olefin content of the
copolymer is usually within the range of from 1 to 10%
25   by mole.

The bag of the invention can be produced by various
techniques. For example, feed resin for the outer layer
and feed resin for the inner layer are melted and kneaded
at usual processing temperature in respective extruders
5   and extruded therefrom, and then both of the resulting
extruded resins are introduced into a circular die
having double slit to form two layers which are bonded
to each other inside the die. Thereafter, the inflation
processing of the bonded two layers is performed at a
10   blow ratio of from 2.5 to 8, preferably from 3 to 6, to
obtain a tubular two layer film from which the bag of
the present invention is produced. The outer layer and
inner layer can be bonded together either outside or
inside the die. It is preferred, however to bond
15   together the layers inside the die since the bonding
strength between the layers can be increased.

In producing the bag of the invention, if desired,
additives such as pigment, a slip agent, an antioxidant,
an antistatic agent and a weather resistance-improving
20   agent may be incorporated into any one or both of the
inner and outer layers at any processing step.

The bag of the invention having the above described
structure is very superior in physical properties such as
film puncture strength and tear strength to the

conventional bags and, furthermore, its seal strength
and impact resistance at low temperature are very high.
Thus the bag of the invention can be used satisfactorily
to accommodate therein and transfer heavy materials,
5   particularly heavy angular materials such as fowl. Moreover,
the bag of the invention is reduced in blocking
properties and has high stiffness and, therefore, it is
suitable for use in automatic packaging and greatly
accelerates packaging efficiency. Furthermore, since
10 the mechanical strength of the bag is great, the
thickness of the bag is sufficient to be about 10 to
200 microns and, therefore, a reduction in thickness can
be achieved, which is economically advantageous.

The invention is illustrated by the following Examples.
15 In these Examples, physical tests were carried out
according to the following methods.

Impact Resistance: Measured using a film impact tester
(manufactured by Toyo Seiki Seisakujo Co. Ltd; specified
impact load: 30 kg.cm; impact hammer diameter 25.4 mm)
20 Puncture Strength: Measured according to JIS-Z-8401.
Tear Strength: Measured according to JIS-Z-1702.
Tensile Modulus: Measured according to JIS-Z-1702.
Tensile Strength: Measured according to JIS-Z-1702.
Elongation: Measured according to JIS-Z-1702.

Seal Strength: Measured according to ASTM-D-1822.

Practical Drop Test: A bag was charged with 20 kg of chemical fertilizer and dropped on a concrete floor from a height of 2.5 meters in such a manner that the broad

5   bag surface was horizontal to the floor. The bag--breakage ratio (number of broken bags/number of dropped bags) was determined.

Processability: By observing the continuous operation condition in processing a two layer film by an inflation

10   method, the processability was evaluated on a numeric scale as follows:

3 ... Excellent, i.e. continuous operation can be performed stably for more than one week.

2 ... Good, i.e. continuous operation can be

15   performed for 1 to 6 days.

1 ... Fair, i.e. continuous operation can be performed for 3 to 24 hours.

Appearance: Evaluated by observing with the eye.

Blocking: Evaluated by examining whether or not the

20   inner surfaces of a bag closely stick to each other in opening the bag after the production thereof.

3 ... The bag can be opened with ease.

2 ... The bag can be opened with slight difficulty.

EXAMPLES 1 to 16

25   Feed resin for an inner layer and feed resin for an outer

layer as shown in Table 1 were melted and kneaded in the
respective extruders and extruded therefrom, and there-
after, both of the resulting extruded resins were
introduced into a circular die having double slit to
5   form two layers which were bonded to each other inside
the die.   Then, inflation processing was performed at a
blow ratio of 4.3 to obtain a tubular two layer film in
which the ratio in thickness of the inner layer to the
outer layer was 4:1 and the total thickness was 80
10   microns.   From this tubular film was produced a bag
having a width of 500 mm and a length of 620 mm wherein
one end of the bag was heat-sealed.   Physical tests
were carried out on the bags, and the results are shown
in Table 1.

15   In Table 1, the plastics materials are identified by
single letters as follows:

   A: High-pressure-produced LDPE having a density of
   0.934 g/cm^3 and a MI of 3 g/10 min.

   B: High-pressure-produced LDPE having a density of
20   0.926 g/cm^3 and a MI of 0.4 g/10 min.

   C: High-pressure-produced LDPE having a density of
   0.921 g/cm^3 and a MI of 0.6 g/10 min.

   D: High-pressure-produced LDPE having a density of
   0.917 g/cm^3 and a MI of 8 g/10 min.

25   E: High-pressure-produced LDPE having a density of

924 g/cm^3 and a MI of 3 g/10 min.

F: Low-pressure-produced LDPE having a density of 0.921 g/cm^3, a MI of 3.7 g/10 min, and a C$_8$ α-olefin content of 10.3% by weight.

5 G: EVA having a density of 0.94 g/cm^3, a MI of 0.6 g/10 min, and a vinyl acetate content of 15% by weight.

H: HDPE having a density of 0.954 g/cm^3 and a MI of 0.05 g/10 min.

10 I: Ethylene-propylene copolymer having a Mooney viscosity ML_{1+4}(100°C) of 60, and a propylene content of 27% by weight.

J: Ethylene-propylene terpolymer having a Mooney viscosity ML_{1+4} (100°C) of 90 and a propylene content of 28% by weight.

15 K: Ethylene-propylene terpolymer having a Mooney viscosity ML_{1+4} (100°C) of 105 and a propylene content of 43% by weight.

L: Ethylene-propylene terpolymer having a Mooney viscosity ML_{1+4} (100°C) of 42 and a propylene content of 43% by weight.

EXAMPLES 17 and 18

The bags of Examples 3 and 4 respectively were charged with 20 kilograms (kg) each of chemical fertilizer, 25 were allowed to stand one day and night in a room

maintained at -20°C, and then were subjected to the

practical drop test to determine the bag breakage ratio.

In Example 17 the bag breakage ratio was 15%, whilst

the bag breakage ratio in Example 18 was 0%.

Table 1

Example	Inner Layer HDPE Type	wt%	EP Type	wt%	Outer Layer LDPE Type	wt%	EVA Type	wt %	Strength of Bag Impact Resistance kg.cm	Puncture Strength kg.cm	Tear Strength MD/TD kg.cm	Tensile Modulus MD/TD kg/cm²	Tensile Strength MD/TD kg/cm²	Elongation MD/TD %
1	H	95	J	5	B	100	-	-	13.0	13.0	22/35	7430/7400	390/400	460/510
2	H	90	J	10	B	100	-	-	13.0	48.0	40/80	6600/6890	380/400	540/500
3	H	80	J	20	B	100	-	-	13.0	96.0	67/110	4810/4960	370/360	540/500
4	H	70	J	30	B	100	-	-	14.0	112.0	90/110	3680/3820	350/360	570/520
5	H	60	J	40	B	100	-	-	15.0	148.0	120/110	3000/3200	340/330	580/550
6	H	50	J	50	B	100	-	-	16.0	165.0	130/110	2500/2700	330/320	580/550
7	H	80	J	20	C	100	-	-	12.0	90.0	50/90	5300/5600	370/430	510/460
8	H	80	J	20	A	100	-	-	11.0	48.0	40/70	5320/5680	370/420	500/450
9	H	80	J	20	E	100	-	-	11.8	48.0	45/70	5300/5700	370/425	500/460
10	H	80	J	20	D	100	-	-	10.0	40.0	40/60	5400/5500	370/390	510/490
11	H	80	J	20	F	100	-	-	11.5	60.0	50/70	5300/5800	370/400	510/490
12	H	80	K	20	A	100	-	-	11.5	50.0	50/70	5320/5680	370/440	500/480
13	H	80	L	20	A	100	-	-	11.0	54.0	60/70	5300/5700	340/390	510/500
14	H	80	I	20	A	100	-	-	11.0	54.0	37/71	5780/5960	360/360	560/620
15	H	80	I	20	B	100	-	-	13.0	60.0	60/90	5120/5330	349/330	540/440
16	H	80	J	20	A	90	G	10	11.0	60.0	45/65	5400/5560	390/430	500/440

Table 1 (continued)

Example	Seal Strength kg.cm	Practical Drop Test (Bag Breakage Ratio) (%)	Processability	Appearance	Blocking
1	2.0	40	3	good	3
2	4.0	15	3	good	3
3	>4.0	0	3	good	3
4	>4.0	0	3	good	3
5	>4.0	0	2	good	3
6	>4.0	0	1	good	3
7	4.0	0	3	good	3
8	4.0	5	3	good	3
9	4.0	5	3	good	3
10	4.0	25	2	good	3
11	4.0	0	3	good	3
12	4.0	0	3	good	3
13	4.0	0	3	good	3
14	3.5	5	3	good	3
15	3.5	5	3	good	3
16	4.0	5	2	good	3

EXAMPLES 24 and 25

Feed resin for an innermost layer, feed resin for an
inner layer, and feed resin for an outer layer as shown
in Table 3 were melted and kneaded in the respective
5   extruders and extruded therefrom, and thereafter, the
three resulting extruded resins were introduced into a
circular die to form three layers which were bonded to
each other inside the die. Then, inflation processing
was performed at a blow ratio of 4.3 to obtain a 50-
10   -micron thick three layer laminated film. The laminated
film thus produced was subjected to the same physical
testings as in Example 1. The results are shown in
Table 3.

In Table 3 the plastics materials are identified by
15   single letters as follows:

P: Ethylene-butene-1 copolymer having a density of
0.921 g/cm^3, a MI of 3.7 g/10 min and a butene-1
content of 7.5% by weight.

Q: High-pressure-produced LDPE having a density of
20   0.924 g/cm^3 and a MI of 3.0 g/10 min.

R: HDPE having a density of 0.954 g/cm^3 and a MI of
0.05 g/min.

S: Ethylene-propylene rubber having a Mooney
viscosity ML_{1+4} (100°C) of 60 and a propylene
25   unit content of 27% by weight.

Table 3

Example	Innermost	Inner Layer R (wt%)	Ethylene-α-Olefin Copolymer (wt%)	Outer Layer	Ratio of Layer Thickness*	Tensile Characteristics Tensile Strength MD/TD (kg/cm²)	Tensile Modulus MD/TD (kg/cm²)	Tensile Elongation MD/TD (%)
24	P	70	P 30	Q	3:6:1	355/420	6000/6500	550/600
25	P	85	S 15	P	3:6:1	350/430	5500/5800	580/600

Table 3 (continued)

Example	Tear Strength MD/TD (kg/cm)	Puncture Strength (kg.cm)	Seal Strength (kg.cm)	Blocking Resistance	Processability
24	30.0/75.0	57.5	6.0	O	O
25	35.0/60.0	60.0	7.0	O	O

* Ratio in thickness of innermost layer to inner layer to outer layer.

CLAIMS:

1.   A packaging bag comprising an outer layer and an
inner layer, characterised in that the outer layer is a
low density ethylene-based polymer or a mixture thereof
with an ethylene-vinyl acetate copolymer, and the inner
5   layer is a mixture of a high density polyethylene and
an ethylene-α-olefin.

2.   A bag according to claim 1 characterised in that
the low density ethylene-based polymer has a density of
10   from 0.91 to 0.94 gram per cubic centimeter and a melt
index of from 0.3 to 10 grams per 10 minutes.

3.   A bag according to claim 1 or claim 2 characterised
in that the low density ethylene-based polymer is an
ethylene homopolymer or a copolymer of ethylene and an
15   α-olefin containing from 3 to 12 carbon atoms.

4.   A bag according to any preceding claim characterised
in that the outer layer comprises from 10 to 95% by
weight of the low density ethylene-based polymer and
from 90 to 5% by weight of the ethylene-vinyl acetate
20   copolymer.

5.   A bag according to any preceding claim characterised
in that the high density polyethylene has a density of

from 0.94 to 0.97 gram per cubic centimeter and a melt index of from 0.01 to 1 gram per 10 minutes.

6. A bag according to any preceding claim characterised in that the ethylene-α-olefin copolymer of the inner layer is an ethylene-propylene-based elastomer.

7. A bag according to any preceding claim characterised in that the inner layer comprises from 95 to 50% by weight of the high density polyethylene and from 5 to 50% by weight of the ethylene-α-olefin copolymer.

8. A bag according to any preceding claim characterised in that the ratio in thickness of the outer layer to the inner layer is from 1/99 to 60/40.

9. A bag according to any preceding claim characterised in that the bag further comprises an intermediate plastics layer between the inner and outer layers.

10. A bag according to any of claims 1 to 8 characterised in that the bag further comprises an innermost plastics layer on the face of the inner layer opposite to the outer layer.

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